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Channel Flow in Partly Solidified Alloy Systems

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ABSTRACT

Channel segregation is a type of macrosegregation which occurs during alloy solidification in situations where a two phase, solid + liquid, 'mushy zone' exists over a large temperature range for a long period of time.

Differences in thermal or solutal expansion or contraction may cause the interdendritic liquid within the mushy zone to differ in density from the bulk liquid and thus provide a driving force for convective flow. The convective flow patterns within the ingot give rise to channels, or streamers, of solute rich material, referred to as 'A' segregates or freckles in the foundry industry, which are left in the ingot after solidification. The defects are observed to run antiparallel to the direction of gravity in the columnar zone of a casting. Although different geometrical configurations are discussed, channels are particularly evident in base chilled configurations where the solute(s) rejected during solidification are less dense than the solvent.

Channel segregation has been examined in Pb alloys containing Sn and/or Sb as well as the transparent $\text{NH}_4\text{Cl-H}_2\text{O}$ system. Channels are shown to originate from liquid perturbations arising from thermosolutal interactions at or near the dendritic interface, and assuming such a model, an analysis of the situation is presented.

KEYWORDS

Solidification, macrosegregation, 'A' segregate, channel, freckle, thermosolutal convection, perturbation, Rayleigh number.

INTRODUCTION

Channel segregation is a special kind of macrosegregation arising from density driven convective flow during alloy solidification. It occurs in situations where a two phase (solid + liquid) mushy zone exists over a large temperature range for an extended period of time. Thermal or solutal, expansion or contraction may cause the interdendritic liquid within the mushy zone to differ in density from the bulk and thus provide a driving force for convective flow.

The remnants of these convective flows are left in the ingot after solidification in the form of channels or streamers of solute rich material which run anti-parallel to the direction of gravity in the columnar zone of a casting.

In large steel billet castings and other situations where heat flow is primarily horizontal through the mold wall they are referred to as 'A' segregates. Figure 1 shows the top section of a 5 ton killed steel ingot (1). Within the columnar zone, near the top, there are channels or 'A' segregates (dark) presumably rich in light elements such as sulphur, phosphorous and carbon. Channel segregation also occurs in situations where heat flow is vertically downward such as in electroslog refined (ESR) and vacuum arc remelted (VAR) ingots as well as directionally solidified Ni-base superalloy turbine blades where they are commonly referred to as freckles. Channel segregation can cause serious problems during subsequent processing, resulting in costly homogenization treatments or rejection of all or part of a cast ingot. Previous work has examined channel formation during directional solidification of base chilled ingots of Pb alloys containing Sn and/or Sb (2-9) as well as the transparent analogue system $\text{NH}_4\text{Cl-H}_2\text{O}$ (4-7,10,11).

Figure 2a shows a vertical section through a Pb-10wt.% Sn ingot which was allowed to partially solidify with the growth direction vertically upward and then quenched from the top to reveal the solidification front. Near the center is a channel of Sn rich material beginning near the bottom and extending to the dendritic interface. Figure 2b is a cross section of a similar ingot at approximately the midpoint showing numerous freckles ~ 1 mm in diameter against a background array of primary dendrite 'crosses'.

The $\text{NH}_4\text{Cl-H}_2\text{O}$ eutectic system has been used for some time as an analogue to study the solidification of metals (12). Due to its low entropy of fusion it



Fig. 1. 'A' segregate channels, rich in sulphur, phosphorus and carbon, in top portion of a 5 ton killed steel ingot (1).

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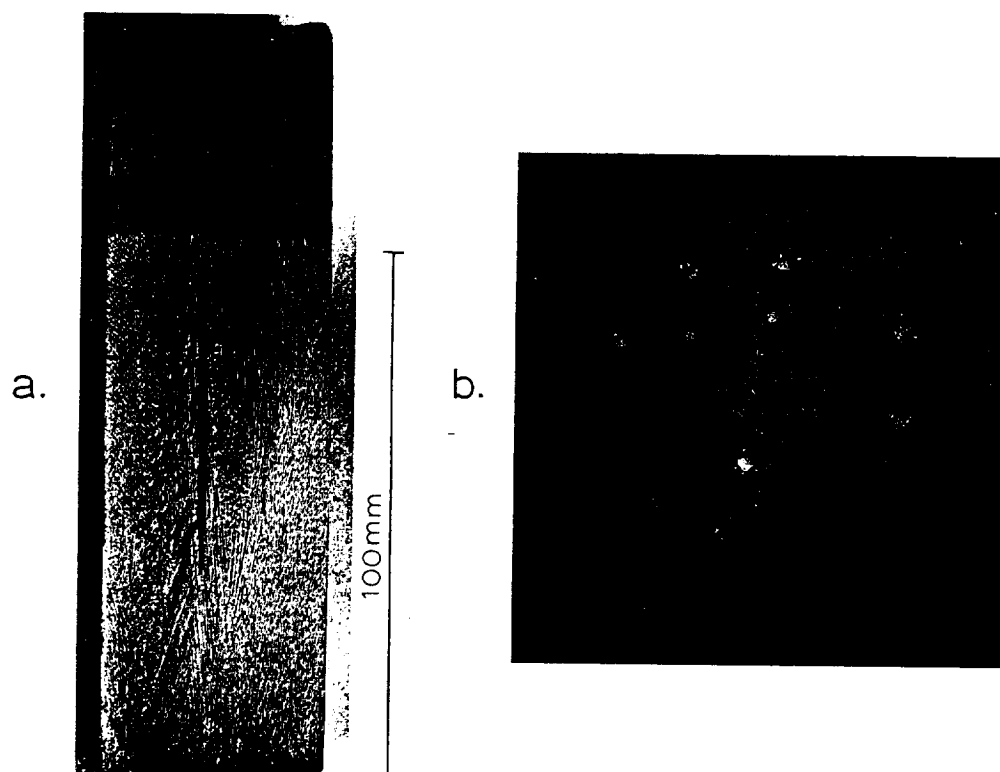


Fig. 2a) vertical and 2b) horizontal sections through a base chilled Pb-10wt.% Sn ingot to show the dendritic growth front and channels.

solidifies dendritically, in a way similar to metallic systems with the distinct advantage of a transparent liquid/opaque solid allowing direct observation of the solidification process. Figure 3a shows a top view of a solidifying NH_4Cl -35 wt.% H_2O casting. It can be seen that channel formation occurs in a similar manner to and on a similar scale as, the metallic systems. In Fig. 3b, a side view of the same casting, a streamlined plume of solute rich liquid can be seen rising from a channel on the dendritic interface. Observation is possible due to a change in refractive index with composition.

Although channels are more clearly found when the solute which is rejected during solidification is less dense than the solvent, similar convective flow patterns may exist during solidification or melting with a solute which is either less or more dense than the solvent. Figure 4.1 represents horizontal heat flow (mushy zone shaded) while Fig. 4.2 and 4.3 represent vertical heat flow parallel and antiparallel to gravity respectively. Figures 4.1-4.3a represent a less dense solute while 4.1-4.3b represent a more dense solute. The flow pattern shown in Fig. 4.1a would be representative of a killed steel ingot (Fig. 1) while Fig. 4.2a reflects the flow pattern observed in the directionally solidified Pb-base or NH_4Cl - H_2O castings (Fig. 2,3). Crystal growth using the Czochralski technique⁴ would exhibit the behavior shown in Fig. 4.3. It should be noted that no convection would be present in Fig. 4.2b since the more dense interdendritic liquid resides stably against the advancing interface.

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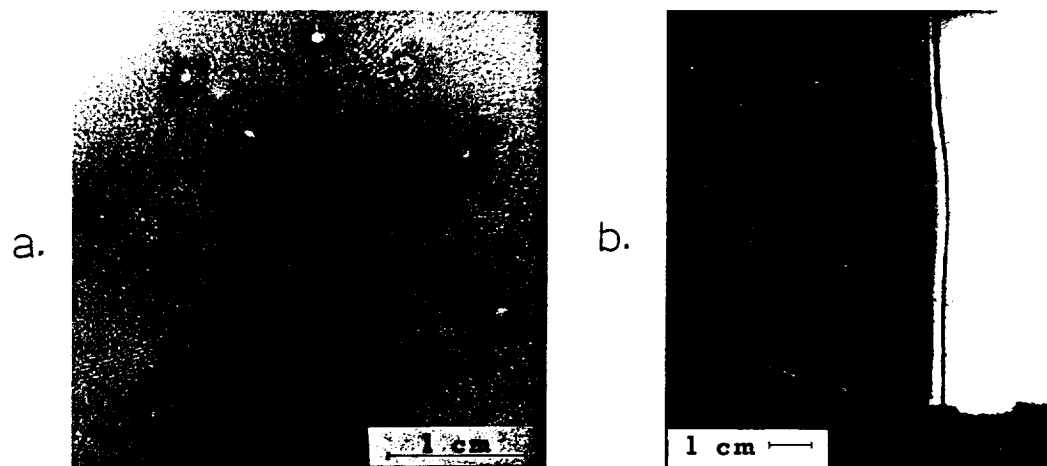


Fig. 3a) Top view and 3b) side view of a solidifying NH_4Cl -35 wt.% H_2O casting observed through the transparent bulk liquid showing channels and plume flow.

Model for Channel Formation

Consider the directional solidification of a base chilled ingot of composition C_0 (Fig. 5, upper left) for the case where the equilibrium distribution is less than 1 and the solute is less dense than the solvent. The solute which is rejected into the liquid during solidification of the primary phase may build up a metastable layer of less-dense, solute-rich liquid in the interdendritic regions, extending a distance of approximately D/V into the bulk liquid, where D is the diffusion coefficient of solute in the liquid and V is the dendrite growth velocity (Fig. 5, upper right).

Examining the concentration profile in the vicinity of the dendritic interface (Fig. 5 lower left) shows a near constant bulk concentration of C_0 , increasing as the interface is approached from the top, due to the presence of the rejected solute. The negative concentration profile (dC/dz) extends down into the mushy zone with a slope dependent on the temperature gradient and liquidus slope until the eutectic front is reached. Since the solute is less dense, the negative concentration profile within the mushy zone results in a positive density gradient (dp/dz) (Fig. 5 lower right). However, the sample is sitting in a positive temperature gradient (dT/dz). As the density decreases slightly with temperature then the density gradient within the bulk liquid (dp/dz) is negative or stabilizing. This results in a metastable condition which may persist for some time.

The formation of channels under these conditions has been well documented since the late 1960's (1,10). However, the origin of the channels or the exact conditions under which they arise, has not been clearly established. Two schools of thought have developed as to where the channels nucleate, one based on theoretical considerations (13,14,15) and the other based on direct experimental observation. The former group contends that channels originate at some imperfection or defect deep within the mushy zone and a channel develops upward by local melting, in a manner similar to a river system. Unfortunately, local acceleration of fluid flow is not possible without cooperative flows



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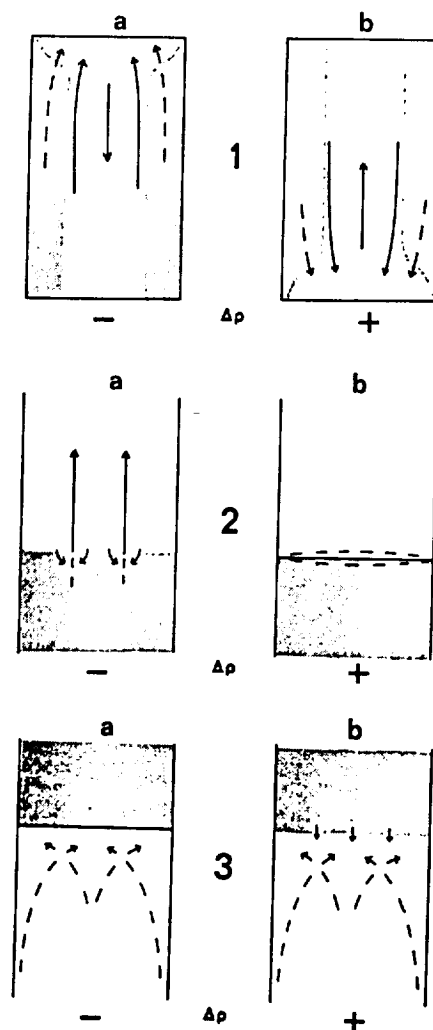


Fig. 4. Convective flow patterns within a casting for alternative heat flow and solute density configurations.

elsewhere within the system. Since the situation is one of a less dense volume of entrapped liquid within the mushy zone surmounted by a body of more dense, bulk liquid, which is thermally stabilized against convection, any motion must originate at or close to the dendritic front where the density gradient changes sign. An analogy has been drawn (11) to the draining of a swamp which is accomplished by creating a breach at the edge rather than by an internal trench or levy.

By direct observation of channel formation in the transparent NH₄Cl-H₂O system, Sample and Hellawell (6) have shown that channels originate from a liquid perturbation at or close to the dendritic interface. A schematic representation of the model is shown in Fig. 6. Figure 6a shows the situation, described previously, that is a less dense boundary layer with a thickness on the order of

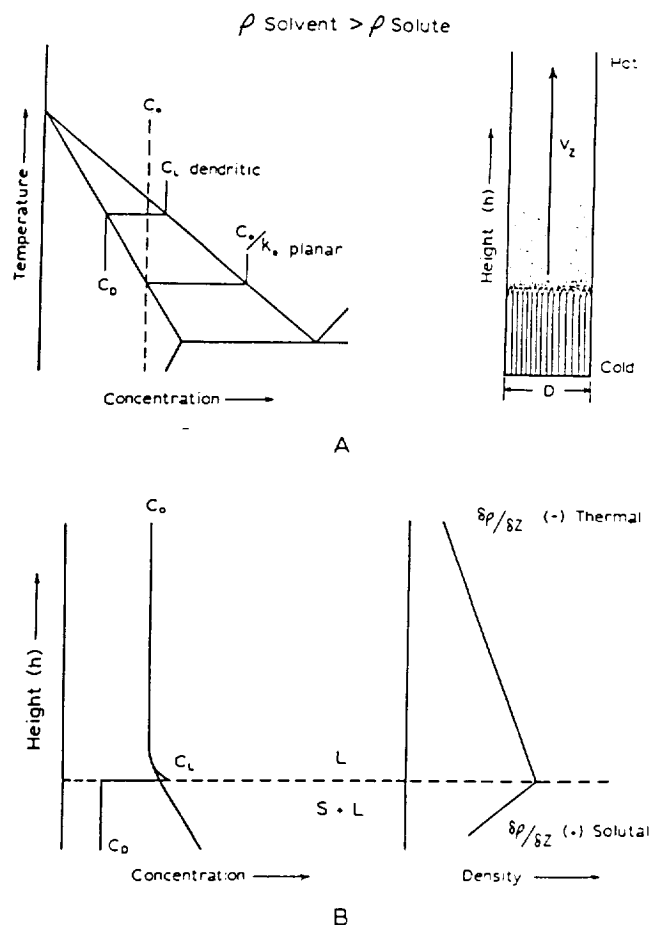


Fig. 5. Schematic concentration/density profiles above and below the dendritic growth front for vertical growth with a less dense solute in a positive temperature gradient.

D/V surmounted by the bulk liquid with the mushy zone below. Since a density inversion exists, the system is metastable and if conditions will allow, it can perturb as shown in Fig. 6b probably by double diffusive or thermal solutal interactions within the boundary layer. As the perturbation grows the concentration gradient and thus the temperature gradient below it is relaxed and the dendrite tips can be observed to accelerate forward to produce a small hillock on the interface as in Fig. 6c. As the perturbation continues to grow it begins to be fed not only by the boundary layer but the interdendritic liquid as well. Upward flow of the interdendritic liquid melts or erodes the dendrites, creating a channel backwards from the interface (Fig. 6d). For channel flow to persist, the upward flow from the channel mouth must be balanced by entrainment of the bulk liquid in the regions immediately adjacent to the channels, Fig. 6e. This results in a quasi-steady state flow pattern which persists until the dendritic interface reaches the top of the casting.

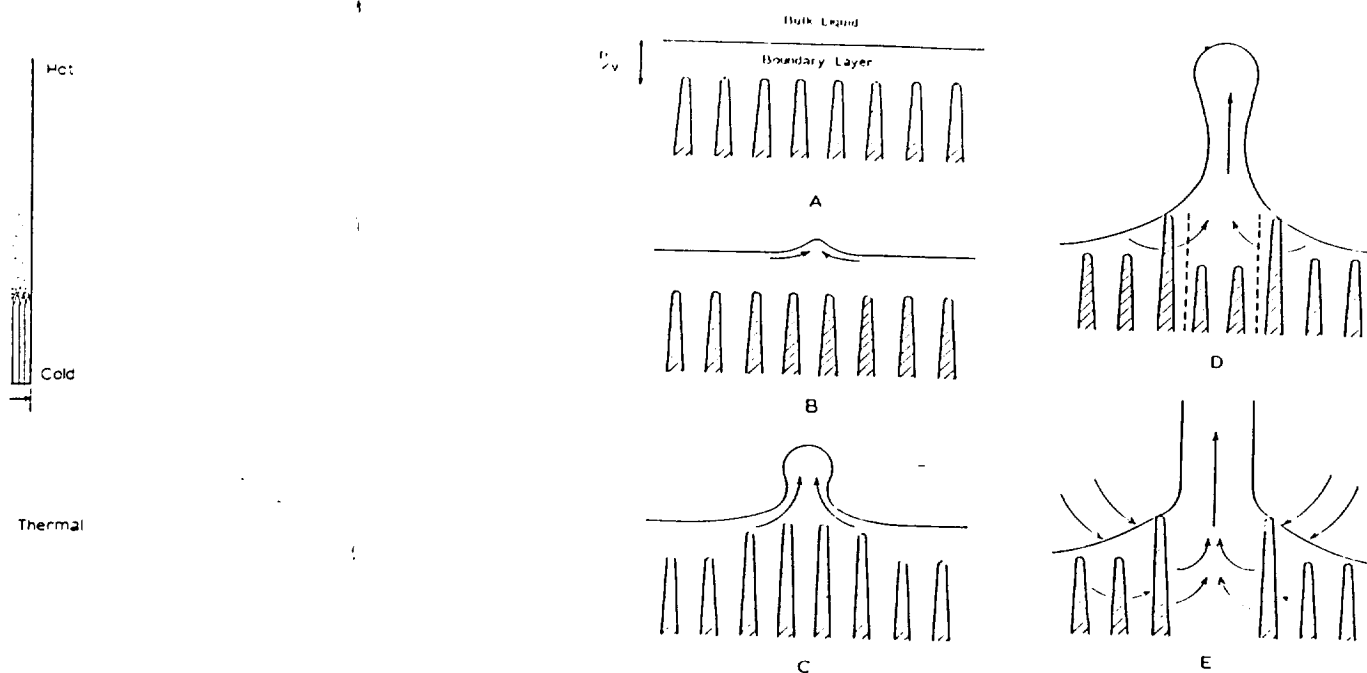


Fig. 6. Schematic model for channel formation by liquid perturbation at the dendritic growth front.

Experimental Work

Although the entire nucleation event occurs within seconds, careful observations of channel formation against the mold wall and simple experiments using the $\text{NH}_4\text{Cl-H}_2\text{O}$ systems (4) have shown that channels nucleate by a liquid perturbation near the dendritic interface and that plume flow precedes channel formation.

Experiments using potassium permanganate (KMnO_4) crystals as dies confirmed the channel flow patterns and local retrainment regions. Attempts were also made to artificially create channels (Fig. 7). An $\text{NH}_4\text{Cl-35 wt.}\% \text{H}_2\text{O}$ casting was allowed to solidify to point where channels were known to have formed. A fine 1 mm bore silica tube, sharpened on the end was then carefully twisted while inserting it into the mushy zone so as to drill out an artificial channel (Fig. 7a). Channels created in this manner failed to propagate and were soon grown over. The experiment was then repeated, this time the tube inserted to a point approximately 1 mm from the dendritic interface and a small amount of liquid drawn up so as to create an artificial plume (Fig. 7b). In each case a channel formed at the exact position where the plume had been created.

Although the $\text{NH}_4\text{Cl-H}_2\text{O}$ system was very convenient to work with for modeling the mechanisms of formation, practical working conditions limited compositions to a narrow range around 35 wt. % NH_4Cl . In order to determine the effect of changing the fraction of solid within the mushy zone, the bottom chilled $\text{NH}_4\text{Cl-H}_2\text{O}$ experiments were repeated using Pb alloys contain Sn and/or Sb (8). The relevant portions of the phase diagrams and regimes of channel formation are shown in Fig. 8.

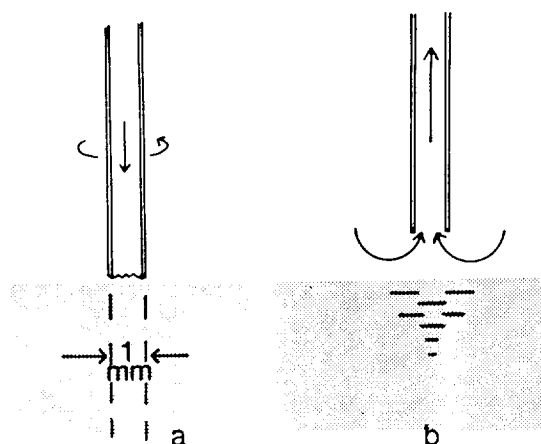


Fig. 7. Artificial formation of channels caused by (a) drilling into the mushy zone and (b) sucking up liquid to create an artificial plume.

In the metallic alloys two regimes of channel formation were observed. Firstly, at low solute contents numerous small channels were observed and secondly, at higher solute contents, a single, centrally located large channel was found (Fig. 9). The transition from the multiple small channels to the single large channel regime has been attributed to the increase in dendritic mesh permeability as the solute content is increased, allowing entrainment patterns on the ingot scale. It should be noted that the regimes of channel formation in the metallic systems are shifted to much higher fractions of solid or lower solute contents than in the analogue system. This is attributed to higher thermal diffusivity and lower kinematic viscosity (Table I) in the metallic system leading to easier establishment of streamlined flow. Within any given ingot in the multiple small channel regime, the number of channels is observed to decrease with distance up the ingot. It can be seen from Fig. 10, a Pb-2 wt.% Sb ingot sectioned at 2 cm intervals from the bottom, that the number of channels decreases with height and every channel present in the top section can be traced back to the bottom. This is consistent with the observations of the analogue castings where channels were observed to coalesce or die out during solidification.

Thermosolutal convection/Rayleigh number

The liquid perturbations leading to the formation of channels have been thought to arise from a phenomenon known as double diffusive or thermosolutal convection (17). This phenomenon is not unique to metallurgy and is believed to be responsible for convective flows in oceanographic, geological or even stellar contexts. Figure 11, after Schmitt (18), is a plot of logarithm of the Lewis number, (Γ), defined as the ratio of the thermal to solutal diffusivity vs. Prandtl number (σ), defined as the kinematic viscosity over the thermal diffusivity, for a variety of double diffusive systems. Systems with high viscosity and low thermal diffusivity fall to the lower right in the figure while those with a low viscosity and high thermal diffusivity tend towards the upper left. The contours represent a normalized flow velocity.

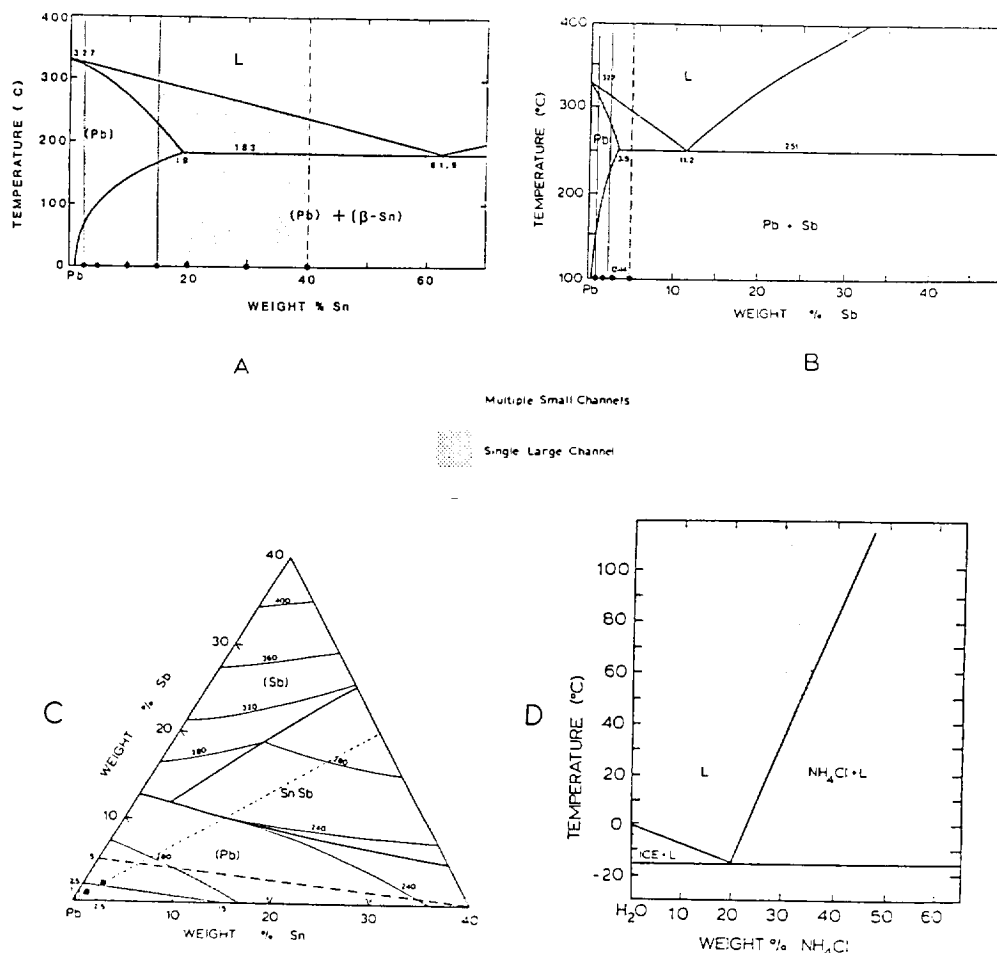


Fig. 8. Partial phase diagrams for (a) Pb-Sn, (b) Pb-Sb, (c) Pb-Sn-Sb and (d) $\text{NH}_4\text{Cl-H}_2\text{O}$ showing compositional regimes (shaded) in which channels were observed.

The phenomenon of thermosolutal convection can arise in any situation where gradients of two or more properties have opposing effects on the vertical density gradient. In this case the properties of interest are heat and solute content. The mechanism of instability can be briefly described as follows: if a small parcel of fluid is displaced upwards its temperature equilibrates quickly but its solute content does not, since the diffusivity of heat is some 2 orders of magnitude greater than that of solute. The fluid parcel is thus solute rich and lighter than its surroundings, becoming increasingly buoyant as it continues to accelerate upwards.

If the solute were to be neglected and the density inversion in the liquid assumed to be due only to a temperature gradient, the classic Benard problem (19) analyzed by Lord Rayleigh in 1916 (20), then one can write a dimensionless thermal Rayleigh number (Ra_m) to describe the initial disturbance or perturbation.

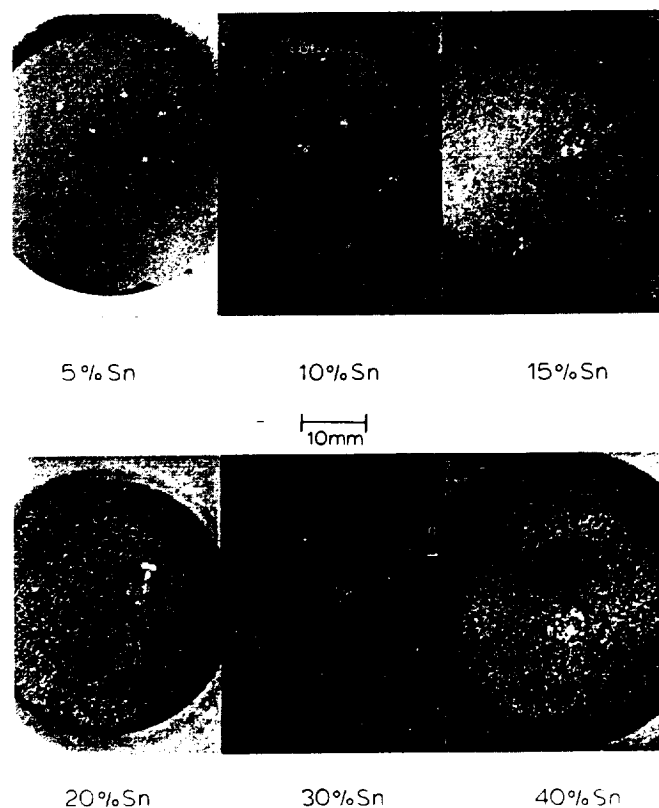


Fig. 9. Horizontal sections of Pb-Sn ingots, 60 mm from the bottom showing change in channel character with increasing concentration.

$$Ra_T = \frac{g \rho \alpha (dT/dz)}{n D_T / h^4} \quad \text{Eq. 1}$$

g = gravitational constant
 ρ = bulk liquid density
 α = volume coefficient of thermal expansion
 dT/dz = vertical temperature gradient
 n = absolute viscosity
 D_T = thermal diffusion coefficient
 h = a characteristic linear dimension

Similarly, if the system were isothermal and the density inversion due only to a solute gradient then an equivalent solutal Rayleigh number (Ra_S) would be:

$$Ra_S = \frac{g \rho \beta (dC/dz)}{n D_S / h^4} \quad \text{Eq. 2}$$

where α is replaced by the volume coefficient of solutal expansion β , dT/dz is replaced by the vertical solute gradient dC/dz and D_T replaced by the mass diffusion coefficient of solute in the liquid D_S .

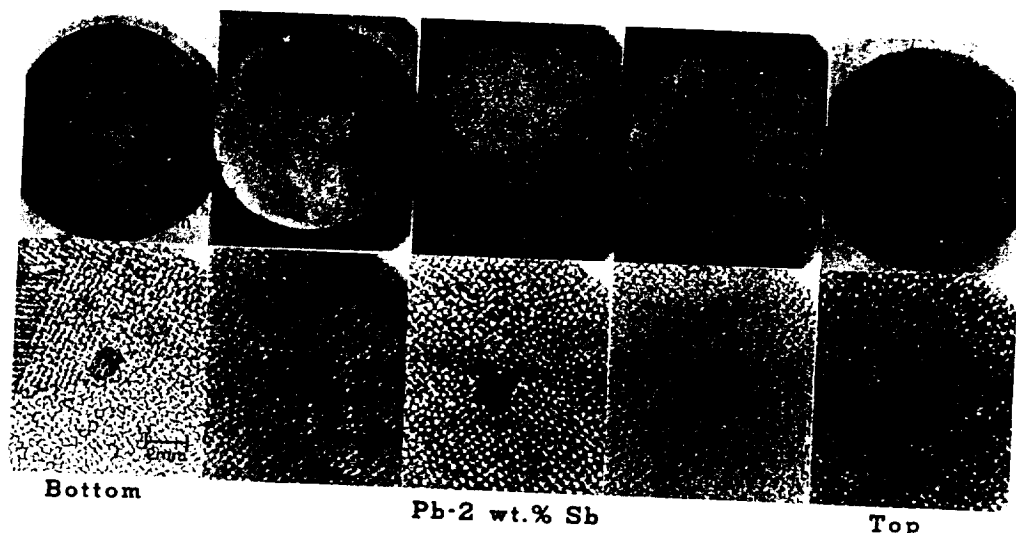


Fig. 10. A Pb-2 wt.% Sb ingot sectioned perpendicular to the growth direction at 20 mm intervals from the bottom.

Closer examination of equations 1 and 2 reveals that the numerators $g\rho\alpha(dT/dz)$ and $g\rho\beta(dC/dz)$ have equivalent units of pressure per unit area (Nm^{-2}). This can be thought of as the buoyant pressure exerted upward due to the density inversion. The units of denominators, $n D_T/h^4$ and $n D_S/h^4$ are the same and this can be thought of as the opposing or restraining pressure due to fluid viscosity and diffusion of heat or solute in the system.

When both thermal and solute gradients are present the Rayleigh numbers can be combined to yield an equivalent thermosolutal Rayleigh number ($Ra_{T/S}$) as follows:

$$Ra_{T/S} = \frac{Ra_S}{\tau} - Ra_T$$

Eq. 3

where τ is the diffusivity ratio or Lewis number. The difference arises since the thermal effect is one of stabilization. $Ra_{T/S}$ may then be written as follows:

$$Ra_{T/S} = \frac{g\rho [\beta(dC/dz) - \alpha(dT/dz)]}{n D_T/h^4}$$

Eq. 4

Analysis

Normally, the system is considered to perturb when $Ra_{T/S}$ exceeds some critical value. However, considerable disagreement exists in the literature as to the value of h , which is alternately taken as D_S/V (21,22), the height (16,24) or the radius (23) of the system. Since h is raised to the 4th power in the equation, these encompass an extremely large range of values making comparison of critical Rayleigh numbers meaningless from case to case.

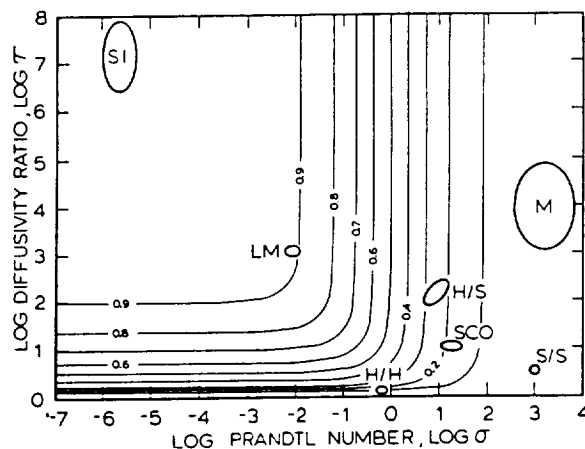


Fig. 11. A plot of Log τ (Lewis number) vs. Log σ (Prandtl number) for a variety of double diffusive systems including liquid metals (LM), aqueous heat/salt solutions (H/S), molten magmas (M), semiconductor oxides (SCO), aqueous salt/sugar solutions (S/S), heat humidity (H/H) and stellar interiors (SI). Contours correspond to a normalized flow rate after Schmitt (18).

In order to determine what might be a reasonable value for h , Sarazin and Hellawell (8,9,25) considered instability to occur when the buoyant forces exceeded the restraining effect of thermal diffusion and fluid viscosity or for a critical effective thermosolutal Rayleigh number ≈ 1 . Back calculation of h from known data for Pb-Sn and $\text{NH}_4\text{Cl-H}_2\text{O}$ revealed h to be on the order of the dendritic spacing in both cases.⁴ The term h appears, therefore, to represent a critical wavelength of a perturbation of the boundary layer. In retrospect it seems entirely reasonable that the perturbation wavelength should be related to the dendrite spacings since a periodic compositional variation would already exist ahead of the advancing dendritic interface, but to arrive at that result independently as it were is quite satisfactory.

The consequences of varying the primary dendrite spacing can be examined for the alloys previously studied. Kurtz and Fisher (26) have given an empirical expression for the dendrite spacing (λ), where:

$$\lambda = \frac{4.3 [\Delta T_0 D \Gamma^{.25}]}{k^{.25} v^{.25} G^{.5}} \quad \text{Eq. 5}$$

- ΔT_0 = temperature difference between liquidus and solidus
- D = mass diffusion coefficient of solute in the liquid
- Γ = Gibbs-Thompson coefficient
- k = equilibrium distribution coefficient
- v = growth velocity
- G = temperature gradient

For a given composition, the primary dendrite spacing is proportional to $v^{-.25} G^{.5}$. Thus, if it were possible to control the thermal gradient and growth velocity, the dendrite spacing and perturbation wavelength for a given alloy could be varied.

Table I
Data for Metallic and Aqueous Systems (4,9,10,16)

	Pb-10 wt.% Sn	Pb-2 wt.% Sb	NH ₄ Cl-35 wt.% H ₂ O
Solvent: solute density ratio (solids)	1.55	1.69	1.53
Liquid density on liquidus, ρ , Kg m ⁻³	9.98 10 ³	~ 10 ⁴	1.08 10 ³
Dynamic viscosity, η , Kg m ⁻¹ s ⁻¹	2.47 10 ⁻³	~ 3 10 ⁻³	1.03 10 ⁻³
Kinematic viscosity, $\nu = \eta/\rho$, m ² s ⁻¹	2.47 10 ⁻⁷	~ 3 10 ⁻⁷	9.54 10 ⁻⁷
Thermal diffusivity, κ , m ² s ⁻¹	1.08 10 ⁻⁵	~ 1 10 ⁻⁵	1.47 10 ⁻⁷
Solutal diffusivity, D , m ² s ⁻¹	3.0 10 ⁻⁹	~ 3.0 10 ⁻⁹	1.3 10 ⁻⁹
Thermal expansion coeff., α , K ⁻¹	1.15 10 ⁻⁴	~ 10 ⁻⁴	6.0 10 ⁻⁴
Solutal expansion coeff., β , wt.% ⁻¹	5.2 10 ⁻³	~ 7 10 ⁻³	2.0 10 ⁻³
Prandtl number, ν/κ	2.3 10 ⁻²	~ 3.0 10 ⁻²	6.81
Lewis number, κ/D	3.6 10 ³	~ 3.0 10 ³	107.7
Fraction liquid at eutectic, f_L	0.079	0.082	0.86
Temperature gradient, dT/dz , K m ⁻¹	10 ³	10 ³	1.5 10 ³
Interdendritic solute gradient, $dc/dc/dz$, wt.%, m ⁻¹	4.3 10 ²	1.5 10 ²	7.0 10 ²
Flow rate to prevent channels, U , m s ⁻¹	10 ⁻³		10 ⁻²
Interdendritic spacing, L , m	3 10 ⁻⁴	3 10 ⁻⁴	5.7 10 ⁻⁴
Derived dimension h , m	6 10 ⁻⁴	4 10 ⁻⁴	4 10 ⁻⁴

Future work will involve assembly of a separately controlled, 2 zone resistance furnace to permit relatively independent control of temperature gradient and growth velocity. This will allow determination of regimes of channel formation on a G-V plot such as the one shown schematically in Fig. 12 (10).

Increasing either the thermal gradient or the growth velocity for a given alloy would decrease the dendrite spacings and thus the perturbations' wavelength, making channel formation more difficult since a larger buoyancy pressure would be required.

Summary

- Under conditions of positive temperature gradient upwards and vertical heat flow downwards, channel segregation has been observed in the transparent analogue NH₄Cl-H₂O system as well as Pb alloys containing Sn and/or Sb.

Eq. 5

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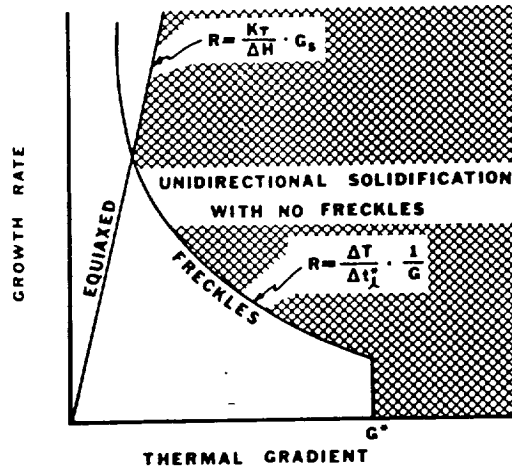


Fig. 12. Schematic plot of growth rate vs. temperature gradient for freckle formation after Copley et al (10).

2. The liquid perturbations leading to the formation of channels arise from thermosolutal interactions at or near the dendritic interface and have been assumed to occur when the thermosolutal Rayleigh number ($Ra_{T/S}$) exceeds 1.
3. The characteristic linear dimension or wavelength (h) in the calculation of Rayleigh numbers is on the order of the dendrite spacings.
4. Dendrite spacings are inversely proportional to the temperature gradient (G°) and the growth velocity (V^{25}).
5. Increasing the thermal gradient or growth velocity for alloys in which channel formation has already been observed, should decrease the dendrite spacings and thus the perturbation wavelength and so make channel formation more difficult since a larger buoyancy pressure would be required.

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